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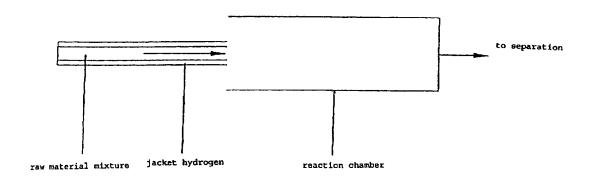
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(54) OXYDES PYROGENIQUES ET PROCEDE DE PREPARATION

(54) PYROGENIC OXIDES AND A PROCESS FOR THEIR PREPARATION

Open mode of operation



- (57) Oxydes pyrogéniques ayant une surface BET comprise entre 30 et 150 m²/g et ayant une viscosité de moins de 2 500 mPas dans une suspension aqueuse. Ils sont préparés en réglant le rapport d'hydrogène gamma et le rapport d'oxygène lambda à moins de l'unité dans une hydrolyse à la flamme à haute température. Les oxydes pyrogéniques sont utilisés pour l'application CMP dans l'industrie électronique.
- (57) Pyrogenic oxides having a BET surface area of between 30 and 150 m²/g have a viscosity of less than 2,500 mPas in an aqueous suspension. They are prepared by setting the hydrogen ratio gamma and the oxygen ratio lambda to less than one in high-temperature flame hydrolysis. The pyrogenic oxides are employed in the CMP application in the electronics industry.

Abstract

Pyrogenic oxides having a BET surface area of between 30 and $150 \text{ m}^2/\text{g}$ have a viscosity of less than 2,500 mPas in an aqueous suspension. They are prepared by setting the hydrogen ratio gamma and the oxygen ratio lambda to less than one in high-temperature flame hydrolysis.

The pyrogenic oxides are employed in the CMP application in the electronics industry.

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Pyrogenic oxides and a process for their preparation

The invention relates to pyrogenic oxides and a process for their preparation and to their use as polishing agents.

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It is known to prepare pyrogenic silicon dioxide and also other metal— or metalloid oxides by means of high-temperature flame hydrolysis (Ullmanns Enzyklopädie der Technischen Chemie [Ullmann's Encyclopaedia of Industrial 10 Chemistry] 4th edition, volume 21 pages 464 et seq.).

In this process, metal and/or metalloid halogen compounds, such as, for example, silicon halogen compounds or organosilicon halogen compounds, are mixed and burned

15 together with (atmospheric) oxygen and hydrogen in a burner of known design. The pyrogen silicic acid formed is then separated off from the gases containing hydrochloric acid, and any chloride still adhering to the oxide is removed by treatment with air containing water vapour.

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The known oxides have the disadvantage that their polishing performance is too low when used for CMP polishing.

There is therefore the object of developing a pyrogenically 25 prepared oxide having an improved polishing performance.

The present invention provides a process for the preparation of pyrogenic oxides by the high-temperature flame hydrolysis process, which is characterized in that the hydrogen ratio gamma (of the raw material gas mixture in the central tube) during the preparation of the pyrogenic oxide is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio lambda (of the raw material gas mixture in the central tube) at the same time also being less than 1, and in particular preferably between 0.7 and 0.9, gamma being the ratio of

hydrogen fed in plus hydrogen from the raw materials to the stoichiometrically required hydrogen and lambda being the ratio of oxygen fed in to the stoichiometrically required oxygen.

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The invention also provides a pyrogenic metal oxide or metalloid oxide, preferably silicon dioxide, prepared by means of the process according to the invention, which is characterized in that during the preparation the hydrogen 10 ratio gamma (of the raw material gas mixture in the central tube) is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio lambda (of the raw material gas mixture in the central tube) at the same time also being less than 1, and in particular preferably between 0.7 and 0.9.

The invention also provides a pyrogenic silicon dioxide prepared by means of the process according to the invention, which is characterized in that the BET surface 20 area of the silicon dioxide is between 30 and 150 m²/g, and a 19 per cent aqueous suspension prepared from the pyrogenic oxide has a viscosity of less than 2,500 mPas, preferably of less than 1,000 mPas.

- 25 The invention also provides a pyrogenic silicon dioxide prepared by means of the process according to the invention, which is characterized in that the BET surface area of the silicon dioxide is between 30 and 150 m²/g and the fractal BET dimension, determined by N₂ adsorption in 30 the pressure range p/p₀ = 0.5 to 0.8 according to the fractal BET theory for multi-layer adsorption by the method described by Pfeifer, Obert and Cole (Proc. R. Soc. London, A 423, 169 (1989)), is less than 2.605.
- 35 Pyrogenic oxides are usually prepared by a method in which the gaseous starting substances are in a stoichiometric

ratio to one another such that the hydrogen, which is either present in the raw materials or fed externally to the combustible gas mixture, is at least sufficient to react the chlorine (from the silicon halogen compounds) present in the combustible gas mixture to give HCl gas. The amount of hydrogen required for this is called the stoichiometric amount of hydrogen.

The ratio of hydrogen fed to the burner (external hydrogen 10 plus chemically bonded hydrogen from the raw materials) to the stoichiometrically required hydrogen just defined is called gamma (y). Accordingly:

 γ = hydrogen fed to the burner / stoichiometrically required amount of hydrogen

or

 $gamma = H_2$ fed in (mol)/ stoichiometric H_2 (mol)

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An amount of oxygen (for example from the air) which is at least sufficient to convert the silicon compounds into silicon dioxide and to convert any excess hydrogen still present into water is furthermore employed in the 25 preparation of pyrogenic oxides.

This amount of oxygen is called the stoichiometric amount of oxygen.

- 30 Analogously, the ratio of oxygen fed to the burner to the stoichiometrically required oxygen is called lambda (λ). Accordingly:
- λ = oxygen fed to the burner / stoichiometrically required amount of oxygen

4

or

lambda = O_2 fed in (mol)/ stoichiometric O_2 (mol)

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The following example is intended to illustrate the use of the terms gamma and lambda:

1 kg SiCl₄ is burned with 0.5 Nm³ hydrogen and 3 Nm³ air.

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The basic reaction equation is:

$$SiCl_4 + 2 H_2 + O_2 ==> SiO_2 + 4 HCl$$

15 2 moles of hydrogen and 1 mole of oxygen are therefore required per mole of SiCl₄.

If 1 kg SiCl₄ (5.88 mol) is reacted, the stoichiometric hydrogen requirement is then 2 x 5.88 mol = 0.263 Nm^3 20 hydrogen.

If 1 kg SiCl₄ is burned with 0.5 Nm³ hydrogen, the calculated gamma is gamma = 0.5/0.263 = 1.9.

- 25 The stoichiometric oxygen requirement is composed of two portions, and in particular on the one hand the portion (a) which is required to form the silicon dioxide, and a second portion (b) to convert excess hydrogen into water.
- 30 The stoichiometric oxygen requirement for the above example is thus calculated as follows:

Amount a): formation of $SiO_2 = 5.88 \text{ mol} = 0.131 \text{ Nm}^3$ (O₂)

Amount b): formation of water from the amount of hydrogen

not reacted with SiCl₄ (in Nm³: 0.5 - 0.263 =

0.237 Nm³ unreacted hydrogen which, according

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to $H_2 + 1/2 O_2 ==> H_2O$, requires an amount of $0.237/2 = 0.118 \text{ Nm}^3 \text{ oxygen.}$

Stoichiometric oxygen requirement = amount a plus amount b $5 = 0.131 + 0.118 = 0.249 \text{ Nm}^3 (O_2)$

If 3 Nm^3 air (with an oxygen content of 21.0 vol.% O_2 , correspondingly = 0.63 Nm^3 oxygen) is employed, the parameter lambda is calculated as follows.

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lambda = 0.63 / (0.131 + 0.118) = 2.53

For raw materials which already contain hydrogen in the molecule, such as e.g. trichlorosilane, the hydrogen

15 contained in the molecule is counted in the calculation as hydrogen additionally fed in.

For molecules which contain carbon in the molecule, when calculating the stoichiometric oxygen requirement it must 20 be taken into account that this carbon must react completely to give CO₂.

A distinction is made between the open and closed mode of operation for the preparation of pyrogenic oxides.

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In the open mode of operation, the reaction mixture burns in a reaction chamber which is open to the atmosphere and in which a reduced pressure prevails, ambient air also being sucked in to the reaction chamber (figure 1).

30

The gaseous mixture of raw materials is premixed homogeneously in a burner of known design and burns in the reaction chamber. To avoid caking, a second annular outflow nozzle, out of which (jacket) hydrogen flows, is placed around the annular outflow nozzle out of which the raw material mixture flows.

According to the definition, in the subsequent considerations and in the calculation of the flame parameters, only the mass streams which are premixed in the central tube are taken into consideration. This means, for example, that the expression "less than stoichiometric" relates only to the ratios in the central tube and not to those in the reaction chamber.

10 In the closed mode of operation (figure 2), the reaction mixture burns in a reaction chamber which is closed off from the atmosphere. In this mode of operation, as a rule an accurately measured amount of secondary air is added in order to avoid the formation of explosive mixtures.

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For normal operation of the reactors for the preparation of pyrogenic oxides, both in the open and in the closed mode of operation, gamma values of greater than 1 (to avoid formation of chlorine) and lambda values of greater than 1 20 (to avoid explosive mixtures) are aimed for.

If an aqueous dispersion is prepared from the pyrogenic oxides, it is found, according to the invention, that during CMP use of these dispersions, the rates of removal of material achieved (for the same specific surface area of the pyrogenic silicon dioxide employed) depend critically on the parameters of gamma and lambda during preparation of the pyrogenic oxide.

- 30 In particular, an increased rate of removal of material by polishing is found if both gamma values of less than 1 and lambda values of less than 1 are established during preparation of the pyrogenic oxide.
- 35 The oxides prepared according to the invention show a significantly increased rate of removal of material during

CMP use compared with known oxides having the same specific surface area, without a deterioration in the surface roughness.

5 This is explained in more detail with the aid of the following examples.

All the following examples were carried out by the open mode of operation (figure 1).

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Methods for determination of the polishing rate and the preparation of the twelve per cent dispersion.

The polishing rate is determined with a dispersion which is twelve per cent with respect to the pyrogenic oxides. The preparation of the dispersions and the method for precise determination of the rate of removal of material are described in WO 95/06690, page 6 in example la. The polishing rate of the twelve per cent dispersion which was prepared from the pyrogenic silicic acid of example 3 is defined as the standard and set equal to one (1).

Since the polishing rate is only determined a fairly long time after the preparation of the pyrogenic silicic acid, a 25 19 % aqueous dispersion is prepared for rapid characterization of the pyrogenic silicic acid. This 19 per cent dispersion is used for monitoring the plant and characterizing the silicic acid.

30 The 19 per cent dispersion is prepared as follows: 38 g pyrogenic oxide and 162 g deionized water are stirred with a dissolver at 2,500 rpm for 5 minutes. The viscosity is measured with a Brookfield viscometer DV 2 at 5 rpm (spindle size 2). The value is determined after one 35 minute.

Example 1: (low rate of removal of material)

2,000 kg/h of a raw material mixture which comprises
silicon tetrachloride to the extent of 84 wt.% and
5 trichlorosilane to the extent of 16 wt.% are vaporized and
the vapour is mixed in a burner of known design with
1,325 Nm³/h air and with 360 Nm³/h of a gas mixture A which
comprises hydrogen to the extent of 94 vol.% and
trichlorosilane to the extent of 6 vol.%, and with
10 177 Nm³/h of a gas mixture B which comprises hydrogen to
the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.%
carbon monoxide and 3.4 vol.% methane. The gas mixture is
ignited, the pyrogenic silicic acid formed is separated off
from the gases and, after treatment with water vapour (for
15 deacidification), the specific surface area is determined
by the BET method. This is 85 m²/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 1.02 and the parameter lambda is calculated as 0.90.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.86.

30 Example 2: (low rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 84 wt.% and trichlorosilane to the extent of 16 wt.% are vaporized and 35 the vapour is mixed in a burner of known design with 1,285 Nm³/h air and with 385 Nm³/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 180 Nm³/h of a gas mixture B which comprises hydrogen to 5 the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for deacidification), the specific surface area is determined 10 by the BET method. This is 85 m²/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is calculated as 1.01 and the parameter lambda is calculated 15 as 0.88.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

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The relative rate of abrasion of the dispersion during polishing is 0.86.

Example 3: (good rate of removal of material)

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2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 96 wt.% and trichlorosilane to the extent of 4 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,100 Nm³/h air and with 281 Nm³/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 229 Nm³/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% 35 carbon monoxide and 3.4 vol.% methane. The gas mixture is

ignited, the pyrogenic silicic acid formed is separated off

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from the gases and, after treatment with water vapour (for deacidification), the specific surface area is determined by the BET method. This is 82 $\rm m^2/g$.

- 5 The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is calculated as 0.92 and the parameter lambda is calculated as 0.77.
- 10 A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during 15 polishing is 1.

Example 4: (good rate of removal of material)

2,000 kg/h of a raw material mixture which comprises
silicon tetrachloride to the extent of 96 wt.% and
5 trichlorosilane to the extent of 4 wt.% are vaporized and
the vapour is mixed in a burner of known design with
1,100 Nm³/h air and with 260 Nm³/h of a gas mixture A which
comprises hydrogen to the extent of 94 vol.% and
trichlorosilane to the extent of 6 vol.%, and with
10 260 Nm³/h of a gas mixture B which comprises hydrogen to
the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.%
carbon monoxide and 3.4 vol.% methane. The gas mixture is
ignited, the pyrogenic silicic acid formed is separated off
from the gases and, after treatment with water vapour (for
15 deacidification), the specific surface area is determined
by the BET method. This is 81 m²/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 0.94 and the parameter lambda is calculated as 0.77.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.99

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Example 5: (good rate of removal of material)

2,000 kg/h of a raw material mixture which comprises
silicon tetrachloride to the extent of 96 wt.% and
5 trichlorosilane to the extent of 4 wt.% are vaporized and
the vapour is mixed in a burner of known design with
1,175 Nm³/h air and with 260 Nm³/h of a gas mixture A which
comprises hydrogen to the extent of 94 vol.% and
trichlorosilane to the extent of 6 vol.%, and with
10 260 Nm³/h of a gas mixture B which comprises hydrogen to
the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.%
carbon monoxide and 3.4 vol.% methane. The gas mixture is
ignited, the pyrogenic silicic acid formed is separated off
from the gases and, after treatment with water vapour (for
15 deacidification), the specific surface area is determined
by the BET method. This is 90 m²/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 0.94 and the parameter lambda is calculated as 0.82.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.98

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Example 6: (good rate of removal of material)

1,900 kg/h of a raw material mixture which comprises
silicon tetrachloride to the extent of 84 wt.% and
5 trichlorosilane to the extent of 16 wt.% are vaporized
together with 100 kg/h propyltrichlorosilane and the vapour
is mixed in a burner of known design with 1,400 Nm³/h air
and with 330 Nm³/h of a gas mixture A which comprises
hydrogen to the extent of 94 vol.% and trichlorosilane to
10 the extent of 6 vol.%, and with 150 Nm³/h of a gas mixture
B which comprises hydrogen to the extent of 90.4 vol.%,
5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.%
methane. The gas mixture is ignited, the pyrogenic silicic
acid formed is separated off from the gases and, after
15 treatment with water vapour (for deacidification), the
specific surface area is determined by the BET method.
This is 86 m²/g.

The flame parameters of the raw materials mixed in the 20 central tube are calculated. The flame parameter gamma is calculated as 0.92 and the parameter lambda is calculated as 0.89.

A 12 per cent aqueous dispersion is prepared from the 25 silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.95

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Example 7: (low rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 96 wt.% and
5 trichlorosilane to the extent of 4 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,125 Nm³/h air and with 350 Nm³/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with
10 250 Nm³/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for 15 deacidification), the specific surface area is determined by the BET method. This is 84 m²/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 1.03 and the parameter lambda is calculated as 0.78.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for 25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.88.

2,000 kg/h of a raw material mixture which comprises

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Example 8: (high rate of removal of material)

silicon tetrachloride to the extent of 84 wt.% and 5 trichlorosilane to the extent of 16 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,125 Nm³/h air and with 320 Nm³/h of a gas mixture A which

comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with

10 125 Nm³/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for

15 deacidification), the specific surface area is determined by the BET method. This is $85~\text{m}^2/\text{g}$.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 0.82 and the parameter lambda is calculated as 0.79.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for 25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 1.04.

30 The reference data of the experiments described are summarized in table 1.

The dependence of the relative rate of removal of material during polishing is plotted as a function of the flame parameters gamma and lambda in figure 3.

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It can be seen that the highest rates of removal of material by the dispersion are found when the flame parameters during preparation of the pyrogenic oxides are significantly under the value 1, and preferably between 0.9 and 0.7.

The different rate of removal of material also manifests itself in the different viscosity of aqueous suspensions which have been prepared from the pyrogenic oxides.

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From table 1 it follows that an inverse relationship exists between the viscosity of the aqueous suspension and the rate of removal of material.

15 The pyrogen silicic acids prepared by the process according to the invention have a reduced fractal dimension compared with the pyrogenic silicic acids prepared by the known process. The reduced fractal dimension is accompanied by an increased rate of removal of material.

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The fractal dimension was determined by N_2 adsorption in the pressure range $p/p_0=0.5$ to 0.8. The measurements results were evaluated according to the fractal BET theory for multi-layer adsorption by the method of Pfeifer, Obert 25 and Cole. (Proc. R. Soc. London, A 423, 169 (1989)).

The dependence of the rate of removal of material on the fractality found is plotted in figure 4. In addition to the regression line, the 95% confidence curve is additionally drawn in there.

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Fractal	dimension				2.607		2.581		2.592	2.601		2.584
Visc.	19%/		[mPas]				400			1,058		52
BET			[b/,ш]		85	85	82	8	06	88	84	85
Rate of	removal	of material	[arbit.	units]	0.88	0.86	1	0.99	0.98	0.95	0.88	1.04
Lambda	core		Ξ		6.0	0.88	0.77	0.77	0.82	0.89	0.78	0.79
Gamma	ലാ		Ξ		1.02	1.01	0.92	0.94	0.94	0.92	1.03	0.82
Mixt. B			[Nm ³ /h]		177	180	229	260	260	150	250	125
Mixt. A			[Nm ³ /h]		360	385	281	260	260	330	350	320
Air	9 20 3		[Nm³/h]		1,325	1,285	1,100	1,100	1,175	1,400	1,125	1,125
PTS			[kg/h]		0	0	0	0	0	100	0	0
Raw material			flow rate	[kg/h]	2,000/R1	2 2,000/R1	3 2,000/R2	4 2,000/R2	5 2,000/R2	6 1,900/R1	7 2,000/R2	8 2,000/R1
Example					1	2	3	4	5	9	7	8

R1 = raw material 1 = 16 wt.% trichlorosilane, 84 wt.% silicon tetrachloride R2 = raw material 2 = 4 wt.% trichlorosilane, 96 wt.% silicon tetrachloride PTS = propyltrichlorosilane
Mixt. A = see text
Mixt. B = see text
Gamma core: hydrogen ratio in the central tube
Lambda core: oxygen ratio in the central tube
Rate of removal of material relative to example 3 (by definition = 1)

Claims

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- 1. Process for the preparation of pyrogenic oxides by the high-temperature flame hydrolysis process, characterized in that the hydrogen ratio gamma (of the raw material gas mixture in the central tube) during the preparation of the pyrogenic oxide is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio lambda (of the raw material gas mixture in the central tube) at the same time also being less than 1, and in particular preferably between 0.7 and 0.9, gamma being the ratio of hydrogen fed in plus hydrogen from the raw materials to the stoichiometrically required hydrogen and lambda being the ratio of oxygen fed in to the
- A pyrogenic metal oxide or metalloid oxide, preferably silicon dioxide, prepared by the process of claim 1, characterized in that during the preparation the
 hydrogen ratio gamma (of the raw material gas mixture in the central tube) is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio lambda (of the raw material gas mixture in the central tube) at the same time also being less than 1, and in particular preferably between 0.7 and 0.9.

stoichiometrically required oxygen.

- 3. A pyrogenic silicon dioxide prepared by the process according to claim 1, characterized in that the BET surface area of the silicon dioxide is between 30 and 150 m²/g, and a 19 per cent aqueous suspension prepared from the pyrogenic oxide has a viscosity of less than 2,500 mPas, preferably of less than 1,000 mPas.
- 4. A pyrogenic silicon dioxide prepared by the process according to claim 1, characterized in that the BET surface area of the silicon dioxide is between 30 and $150~\text{m}^2/\text{g}$ and the fractal BET dimension, determined by N_2

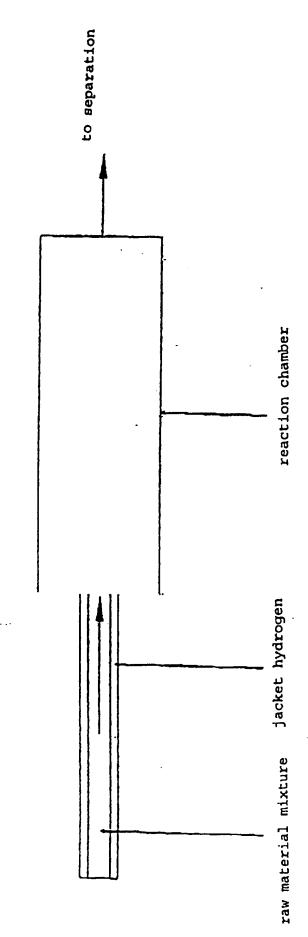
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adsorption in the pressure range $p/p_0=0.5$ to 0.8 according to the fractal BET theory for multi-layer adsorption by the method described by Pfeifer, Obert and Cole. (Proc. R. Soc. London, A 423, 169 (1989)), is less than 2.605.

- 5. Use of the pyrogenic oxides prepared by the process according to claim 1 as a raw material for the preparation of dispersions which are employed for CMP applications.
- 6. Use of the pyrogenic oxides prepared by the process according to claim 1 as a raw material for the preparation of dispersions which are employed for polishing applications in the electronics industry.

Open mode of operation

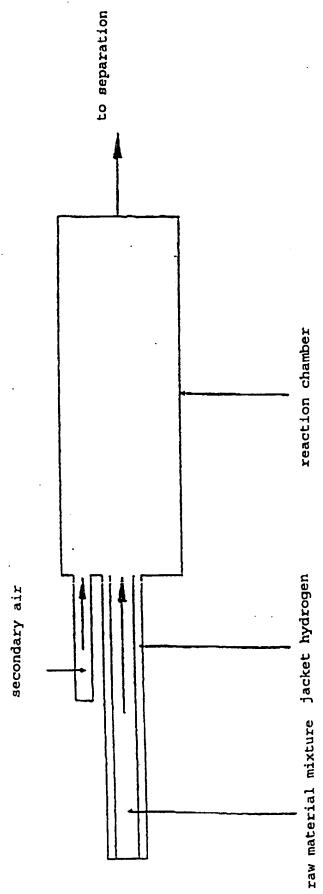
Figure 1



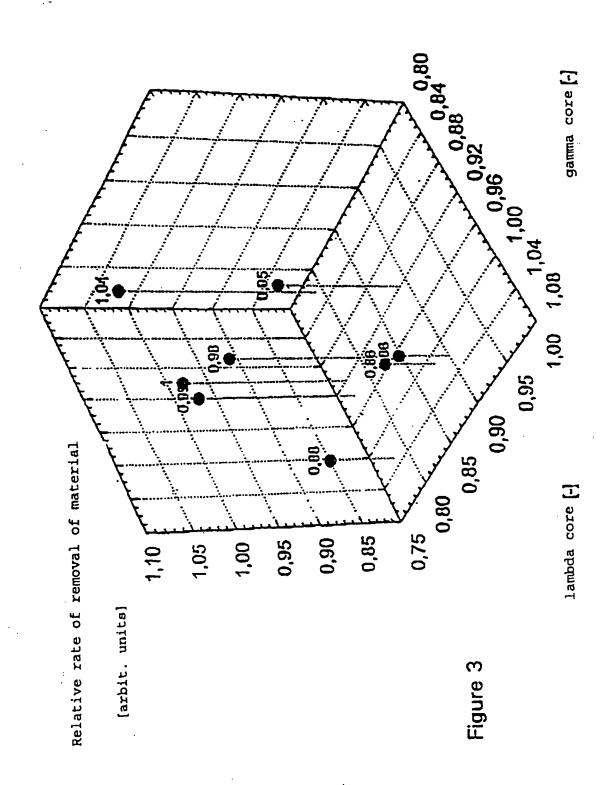
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"Closed" mode of operation

Figure 2



Rosels & Clark



Made Wet

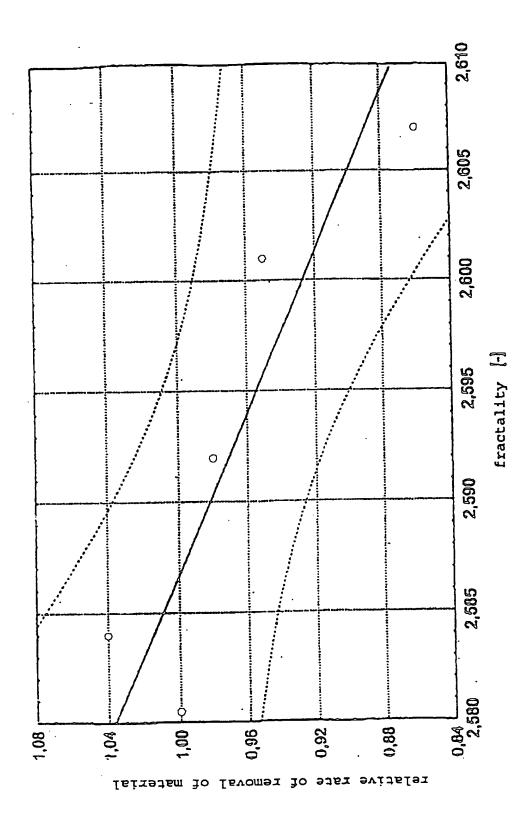


Figure 4

Marks a deals